Energy-Synchronized XAS-PEEM Platform for Nanoscale Magnetic Characterization

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Abstract : This study develops an integrated X-ray Absorption Spectroscopy (XAS)-Photoemission Electron Microscopy (PEEM) platform at the Shanghai Synchrotron Radiation Facility (SSRF) BL09U, enabling nanoscale characterization of complex materials through energy-resolved imaging and micro-zone XAS. By leveraging SSRF's energy/polarization tunability and PEEM's surface sensitivity, we established a GAP-MONO control system under the EPICS framework to dynamically synchronize elliptically polarized undulator (EPU) gap and monochromator energy, optimizing photon flux stability for absorption fine structure analysis. Combining X-ray Magnetic Circular Dichroism (XMCD) /Magnetic Linear Dichroism (XMLD) with PEEM and micro-zone XAS, this platform achieves concurrent mapping of electronic structures and magnetic domains in ferromagnetic nano-patterns, as demonstrated through our $Ni_{80}Fe_{20}$ Permalloy studies based on this system. The dual-modal approach bridges synchrotron radiation technology with surface science, offering nm-scale spatial resolution in XAS with magnetic domain sensitivity through circularly polarized X-ray excitation, providing researchers with advanced tools for functional material analysis through synergistic XAS-PEEM techniques and dynamic control systems.

.1. Introduction

The BL09U "Dreamline" at the Shanghai Synchrotron Radiation Facility (SSRF) is a soft X-ray beamline boasting a broad energy range and exceptionally high energy resolution[1]. Its energy spectrum spans from 20 eV to 2000 eV, with an energy resolution reaching up to 35,000 at 867 eV. Presently, the beamline offers three experimental end-stations for users: The Angle-Resolved Photoemission Spectroscopy (ARPES), the Photoemission Electron Microscopy (PEEM), and the Resonant Inelastic X-ray Scattering (RIXS).

PEEM technique operates based on the photoelectric effect [2,3], where sample surfaces are excited by light to emit electrons that are directly imaged through electromagnetic lenses, enabling full-field nanoscale imaging. This method provides atomic-scale spatial resolution, making it

uniquely capable of resolving lateral heterogeneities in chemical states, magnetic structures, and thin-film interfaces. However, its detection depth remains confined to only few atomic layers due to electron inelastic mean free path limitations. In terms of excitation source selection, while traditional electron beams, lasers, and UV light sources remain in use, synchrotron radiation has emerged as the preferred choice in modern research. Synchrotron X-rays exhibit superior characteristics including extreme brightness, high collimation, and full polarization control, which collectively enhance PEEM's analytical capabilities for fundamental studies. The technique's advantages in rapid imaging and spatial resolution have driven its widespread adoption across multidisciplinary fields including life sciences, materials science, thin-film growth, chemical reactions, catalysis, etc. Notably, PEEM's synergistic integration with complementary techniques has expanded its applicability. When combined with X-ray absorption spectroscopy (XAS) and its derivative techniques, as well as with magnetic circular and linear dichroism technologies, PEEM exhibits broad application prospects in the fields of chemistry, ferromagnetic and antiferromagnetic materials, and electronic structures.

XAS is a powerful analytical technique for probing the local electronic structure and chemical state of materials [4,5], providing critical insights into valence states and electronic environments that form the basis for understanding material properties. The technique operates by exposing materials to highly monochromatic X-ray beams, which generate multiple detectable signals through absorption, reflection, or transmission phenomena. Specifically, transmitted or reflected X-rays produce absorption spectra, while excited Auger electrons and fluorescence photons enable surface-sensitive chemical analysis and depth profiling of electronic structures, respectively. This multi-signal capability allows concurrent characterization of elemental composition, surface chemistry, and bulk electronic properties. Its versatility has established it as an indispensable tool across disciplines, particularly in material science (e.g., catalyst characterization), surface chemistry (e.g., interface reaction monitoring), and nanotechnology (e.g., quantum dot electronic mapping), etc.[4,5].

In XAS research, X-rays in soft-X ray range play a important role in the near-edge absorption spectrum, primarily corresponding to the K edges of carbon and oxygen, and the L/M/N edges of many transition metals. These characteristic energy levels enable distinct analytical advantages: carbon/oxygen edges provide novel insights into organic and carbon-based materials through valence state mapping, while transition metal edges serve as fundamental probes for studying chemical composition and magnetic properties in metals and metal oxides. The complementary nature of these spectral regions allows researchers to simultaneously investigate both surface chemistry and bulk electronic structures in complex materials.

However, traditional XAS techniques face limitations in resolving microscopic features due to their macroscopic measurement scope. X-PEEM overcomes this by integrating high spatial resolution and sensitivity, enabling micro-zone XAS analysis. This is achieved through detecting photon energy-dependent variations in electron emission (both photoelectrons and secondary electrons), which directly correlates with elemental absorption characteristics. The combined advantages of X-PEEM allow concurrent chemical state mapping and magnetic domain analysis at the micro-scale, expanding its applicability to localized property studies in advanced materials.

The micro-zone XAS technique, integrating the strengths of synchrotron radiation sources and photoemission electron microscopy, enables high-resolution XAS measurements at micron and even nanometer scales. The advancement of this technology not only broadens the application

scope of XAS but also offers novel perspectives and tools for the investigation of microstructures and properties in materials science. In recent years, with continuous improvements in data processing algorithms and imaging technologies, the micro-zone XAS technique is poised to seize new developmental opportunities, promising to play an increasingly pivotal role in the fields of materials science, chemistry, and physics.

2．Description of the Experimental System

The experimental system encompasses an X-ray source with adjustable energy and polarization, a photoemission electron microscope, and an advanced data acquisition system.

2.1 Tunable X-ray Source with Adjustable Energy and Polarization

The Dreamline at SSRF utilizes dual APPLE II-type elliptical polarization undulators [6,7], enabling seamless energy regime switching between 20–200 eV (Low Energy EPU, LEID) and 200–2000 eV (High Energy EPU, HEID) while maintaining stable electron beam orbits. X-ray energy and polarization states are precisely tunable via GAP/shift parameter adjustments [8]. In order to meet the diverse experimental requirements for energy resolution while achieving an exceptional energy resolution of up to 35,000@867eV, the system incorporates a variable included angle plane grating monochromator [9] with four gratings: low-energy (400 l/mm LEG), medium-energy (800 l/mm MEG), high-energy (1200 l/mm HEG), and very-high-resolution (3600 l/mm VEG). This multi-grating configuration achieves wide X-ray beam energy across 20–2000 eV, supporting diverse experimental demands while enabling precise characterization of material electronic structures and chemical states through synchrotron-based X-ray spectroscopy.

2.2 Photoemission Electron Microscope

The PEEM station at the SSRF (shown in Figure. 1) incorporates the LEEM-III system from Germany's Elmitec [10,11], enabling comprehensive material science research through integrated imaging, diffraction, and photoelectron spectroscopy capabilities. The system operates based on electron excitation and collection mechanisms: when samples are irradiated by light sources (femtosecond lasers, UV, or synchrotron radiation) or electron beams, emitted electrons are collected by objective lenses, accelerated through electron optical elements, and projected onto detectors to form magnified images [12]. This process simultaneously achieves high-resolution surface morphology imaging and dynamic electron emission analysis under various excitation conditions.

Beyond conventional Low Energy Electron Microscopy (LEEM) functionality, the station supports multiple advanced imaging modes including Micro-Electron Microscopy (MEM), Ultraviolet PEEM, X-ray PEEM, and laser-PEEM imaging modalities [12]. The X-PEEM mode particularly excels in resolving surface fine structures and compositions through synchrotron X-ray excitation, offering element-specific imaging via energy tuning to match absorption edges. This enables precise elemental mapping with enhanced specificity. Additionally, the facility integrates micro-zone X-ray Photoelectron Spectroscopy (XPS) and ARPES techniques for analyzing elemental composition, chemical states, and electronic structures at microscopic scales.

In this work, we expanded the experimental capabilities by developing energy-resolved

PEEM imaging, micro-zone XAS, and X-ray Magnetic Circular Dichroism (XMCD) /Magnetic Linear Dichroism (XMLD) techniques. By leveraging synchrotron radiation's tunable energy and polarization properties, we established advanced methodologies for correlated electron-magnetic-structural characterization, significantly enhancing the experimental station's versatility and research impact in materials science.
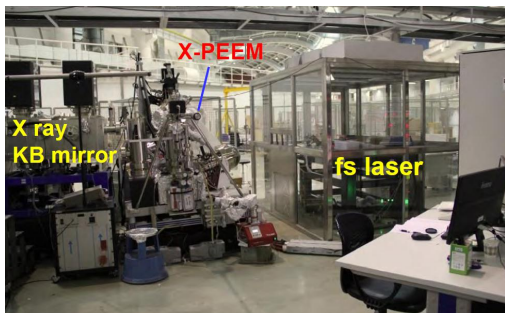

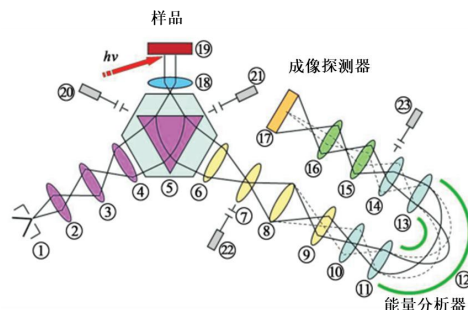Figure 1. Photo of the PEEM End-station at SSRF   Figure 2. Schematic of the Electron Optics in PEEM

2.3 Implementation of Energy-Resolved PEEM Imaging and Micro-zone XAS Techniques

First, Synchrotron radiation X-rays are focused onto the sample surface to generate photoelectrons, which are collected by the PEEM system to produce images revealing local variations in photoemission intensity. Energy-resolved PEEM imaging is achieved by systematically adjusting the monochromator's reflecting angle to select specific X-ray wavelengths, acquiring PEEM images at each discrete energy value.

The SSRF beamline control system utilizes the Experimental Physics and Industrial Control System (EPICS) [13]. To enhance experimental efficiency near elemental absorption edges, we developed a LabView-based automated data acquisition software that integrates with EPICS through a dedicated interface library, enabling concurrent control of the EPU, monochromator, and camera systems. This software automates the workflow by configuring camera exposure parameters, initializing monochromator energy positions, and sequentially capturing single-energy images across the target energy range. By moving the monochromator stepwise and repeating image acquisition, the system rapidly generates a comprehensive PEEM image series near the absorption edge of the element under study.

This integrated approach not only streamlines energy-resolved imaging but also ensures precision in multi-device synchronization, significantly improving experimental throughput. The resulting high-resolution image dataset forms a robust foundation for advanced analysis of electronic structures and chemical states in materials science research.

The energy-resolved PEEM imaging system not only acquires high-resolution images but also synchronously collects photoelectron yield data from the interrogated regions. By analyzing the energy-dependent variation in total photoelectron yield, we reconstruct the sample's absorption spectrum, revealing its electronic structure response to different excitation energies. Furthermore, the integrated micro-region analysis capability enables precise targeting of specific areas for localized yield mapping, thereby obtaining spatially resolved absorption spectra through XAS at the microscopic scale. This dual functionality - combining macroscopic spectral characterization with micro-zone spectroscopic analysis-expands the application of XAS beyond traditional bulk material studies to complex nanostructured systems. The automated data processing pipeline, which integrates real-time yield data acquisition with spectral reconstruction algorithms, provides

a comprehensive toolkit for investigating material properties such as optical characteristics, chemical states, and magnetic domain distributions in advanced research domains including surface chemistry, nanotechnology, and condensed matter physics.

2.4 XMCD/XMLD for Studying the Micro-Structures of Magnetic Materials

Based on the above spectroscopic XMCD and XMLD was further established. Both dichroism methods are used to probe the electronic configuration modifications in magnetic systems, manifested through energy-dependent intensity variations in the near-edge X-ray absorption fine structure. The observed spectral changes arise from the interplay between the material's magnetic moments and the incident X-ray polarization, specifically dependent on the magnetic moment's magnitude and orientation [14,15].

Experimentally, we employed $Ni_{80}Fe_{20}$ Permalloy nanopatterns prepared on Si substrates through magnetron sputtering for PEEM studies. The 70-µm diameter imaging field contained arrays of $Ni_{80}Fe_{20}$ disks exhibiting vortex magnetic domain structures, with diameters varying between 300 nm and 2400 nm and uniform thickness (~50 nm). To elucidate the electronic and magnetic characteristics of these nanostructures, we conducted energy-resolved PEEM measurements near the Fe L-edge (700-730 eV) using circularly polarized X-rays with 0.2-eV energy resolution. For each energy level, we determined the spatially averaged photoelectron intensity across the entire patterned area, thereby constructing the element-specific absorption spectrum as illustrated in Figure 3(e). This approach simultaneously provides both morphological information about the magnetic domains and quantitative insights into the material's electronic state evolution.
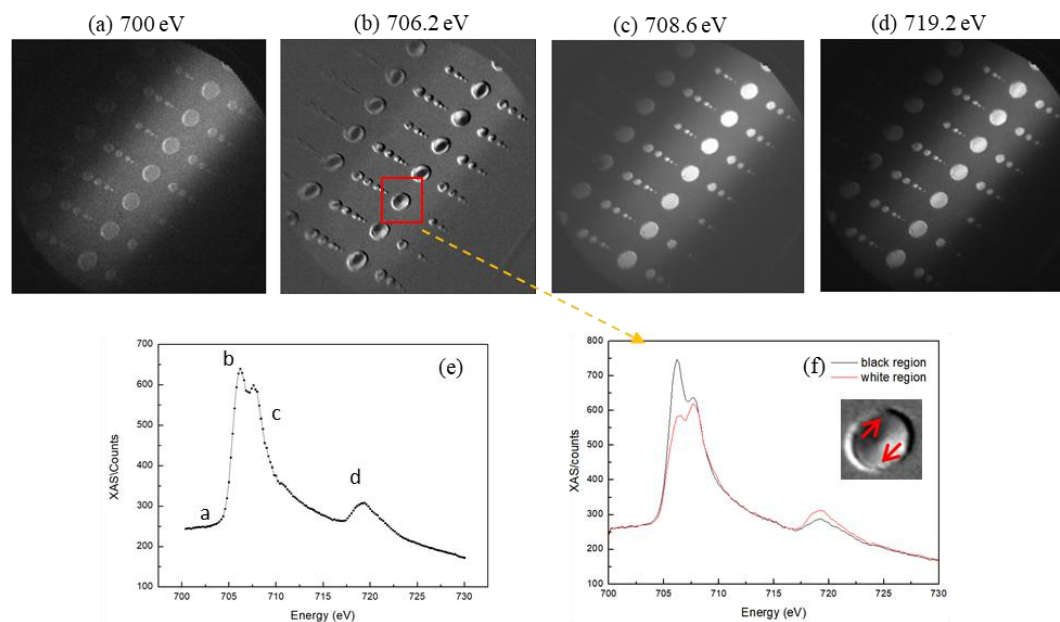
Figure. 3. PEEM images taken at energies of (a) 700 eV, (b) 706.2 eV, (c) 708.6 eV, and (d) 719.2 eV. (e) The absorption spectrum for the whole imaged region. (f) Absorption spectra of two micro-zones with different contrasts in the area enclosed by a red box in (b).

Figure 3(a)-(d) displays the energy-dependent PEEM imaging results of $Ni_{80}Fe_{20}$ nano-disks at 700 eV, 706.2 eV, 708.6 eV, and 719.2 eV. These images reveal quantitative intensity contrasts corresponding to variations in the photoemission yield across the magnetic domains. Notably, the

Fe L_3-edge (706.2 eV) and L_2-edge (719.2 eV) images (Figures 3b and 3d) exhibit pronounced vortex-like magnetic domain structures characterized by distinct bright/dark contrast regions. The enhanced intensity modulation at 706.2 eV (Figure 3b) facilitates detailed morphological analysis of these magnetic vortices.

To investigate the electronic origins of the observed contrast variations, we performed micro-spectral mapping on a representative vortex from Figure 3b. Figure 3f compares the absorption spectra of two regions with contrasting intensities: one exhibiting bright emission (high-intensity region) and the other showing dark emission (low-intensity region). The spectral data demonstrate significant differences in the Fe 3d electronic states between these micro-regions, directly correlating with their distinct magnetic moment configurations.

By analyzing the circularly polarized X-ray induced transitions at the Fe L_3 ($2p_{3/2} \rightarrow 3d$) and L_2 ($2p_{1/2} \rightarrow 3d$) edges, we establish a direct relationship between the electronic structure evolution and the spatial distribution of magnetic domains. The energy-selective imaging approach not only provides morphological characterization of the vortex structures but also enables quantitative analysis of the spin-dependent electron distribution within the nano-patterned material.

2.5 GAP-MONO Coupled XAS Method

To address the need for high-quality data acquisition in the field of fine structure studies of absorption spectra, this work is dedicated to optimizing experimental conditions to ensure that the photon flux, used as a normalization reference, reaches its maximum at every energy point within the scanning range. Achieving this goal hinges on accurately correlating the EPU GAP with the X-ray energy, thereby enabling the coordinated adjustment of the EPU GAP and energy (GAP-MONO linkage).

Building on the previously established data acquisition software for the energy-resolved PEEM imaging system, we developed enhanced control interfaces between the experimental station and the EPICS framework. This integration allows the system to automatically synchronize the EPU gap with monochromator energy settings, thereby implementing a GAP-MONO-linked micro-zone XAS technique. For soft X-ray elements exhibiting absorption spectra spanning tens of eV, we observed a quasi-linear relationship between maximum photon flux and X-ray energy. Based on this empirical observation, we established the linear regression model: $GAP = K*E+b$, where K and b are calibration coefficients determined by measuring gap values at the Start/Stop energy points before experimentation.

Experimental validation demonstrates the effectiveness of this approach through comparative measurements of the STO sample's oxygen edge spectrum in coupled versus uncoupled modes. Photon flux data, derived from the upstream gold mesh photocurrent, shows significant improvement across the scan range when using the GAP-MONO linkage method. This optimization not only stabilizes the normalization reference but also enhances spectral resolution, providing researchers with superior conditions for analyzing fine structural features in absorption spectra.
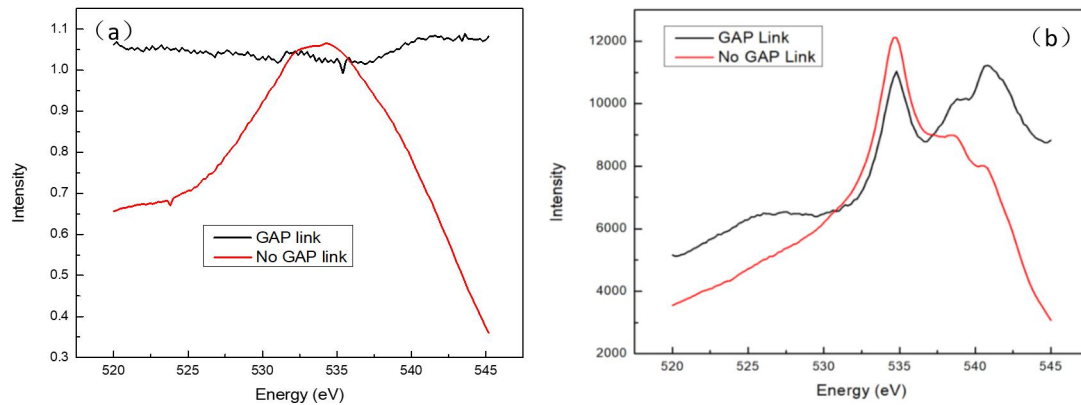
Figure. 4. Comparison of photon flux (a) and absorption spectra (b) in coupled and uncoupled modes.

3. Conclusion

In summary, this work establishes a comprehensive experimental tool combining the tunable energy and polarization capabilities of the PEEM endstation at BL09U "Dreamline" in SSRF with advanced energy-resolved PEEM techniques. By integrating XMCD/XMLD and micro-zone XAS, we achieve spatially resolved electronic structure mapping and magnetic property analysis at the nanoscale level. This dual-modal approach enables precise identification of optical properties in complex materials, providing powerful tools for frontier research in condensed matter physics and nanotechnology. Furtherly, in order to meet the needs for stable photon flux control in absorption spectrum fine structure studies, we developed a GAP-MONO linkage that dynamically EPU gap adjustments with monochromator energy settings. Through linear regression modeling based on calibration measurements at energy extremes, this innovation achieves real-time optimization of photon flux stability across extended energy ranges. The developed system demonstrates superior performance in resolving fine spectral features, as validated through our $Ni_{80}Fe_{20}$ Permalloy nanopattern studies.

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